

Photon lasing in a GaAs microcavity: similarities with a polariton condensate

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We study experimentally the lasing regime of a GaAs based microcavity sample under strong optical pumping. The very same sample exhibits the strong coupling regime at low excitation power with a Rabi splitting as large as 15 meV. We show that some features which may be considered as experimental evidence of polariton Bose Einstein condensation are also observed in the weak coupling regime when the cavity is behaving as a regular photon laser. In particular, the emission pattern in the lasing regime displays a sharp peak near the energy minimum followed by a Boltzmann distribution at higher energies.

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Microcavity polaritons are the quasi-particles arising from the strong coupling regime between a cavity mode and quantum well excitons¹. Because of their bosonic nature, microcavity polaritons are expected to undergo a Bose-Einstein condensation with the appearance of spontaneous coherence^{2,3}. The polariton dispersion presents a pronounced energy trap close to the center of the Brillouin zone⁴. As a result, polaritons exhibit a very small effective mass ($\sim 10^8$ smaller than the hydrogen atom mass) and thus are expected to condensate at unusually high temperatures (up to room temperature in wide band gap microcavities⁵).

Claims of polariton condensation have been published these last years by several groups in different cavity geometries and in different semiconductor systems. In planar cavities, strong evidence for the spontaneous appearance of polariton spatial coherence has been obtained in II-VI CdTe based samples⁶ at low temperature (typically 10 K). More recently, polariton lasing has been claimed at room temperature in wide band-gap GaN based microcavities⁷ but definite proof of the persistence of the strong coupling regime in these measurements still needs to be established. Concerning planar GaAs based microcavities, it is now well established that polariton lasing cannot be obtained under high energy non resonant excitation^{8,9,10,11}: because of a relaxation bottleneck^{12,13}, polaritons do not scatter efficiently enough toward the lowest energy states and eventually the strong coupling regime is bleached before polariton state occupancy in the energy minimum reaches unity. To circumvent this inefficient polariton thermalization, "cold" excitation has been successfully used by directly injecting polaritons on the high energy states of the lower polariton branch¹⁴. Finally very recently Babili et al.¹⁵ reported on a strong non-linear coherent emission in a GaAs based microcavity under high energy non resonant excitation using a spatially localized energy trap induced by strain.

Because at high excitation power, conventional lasing (VCSEL) can occur in a semiconductor microcavity, it is delicate to ensure that the observed emission is actually due to polaritons, especially in new materials or when introducing a new geometry.

In the present work, we want to underline that striking similarities exist between a polariton laser and a standard VCSEL. We perform emission measurements under high energy non resonant excitation in a large Rabi splitting sample analogous to the one used in Refs. 14 and 15. Strong non-linear emission is observed which is shown to be due to electron-hole pair lasing by monitoring the in-plane dispersion of the emission. Nevertheless we show that the lasing emission does not always occur at the expected energy of the bare cavity mode but it is significantly redshifted. Moreover, monitoring the emission as a function of energy, we find an intensity distribution completely similar to that reported for a polariton laser. Therefore, these experimental features cannot be used on their own as a proof for the establishment of a polariton condensate in coexistence with a thermalized polariton cloud.

Our sample, grown by molecular beam epitaxy, consists in a $\lambda/2$ AlAs cavity surrounded by two $Al_{0.2}Ga_{0.8}As/AlAs$ Bragg mirrors with respectively 16 and 20 pairs on the top and bottom mirror. To get a large Rabi splitting with a small cavity volume, we inserted quantum wells not only in the cavity layer but also at the first antinodes of the electromagnetic field in each Bragg mirror as firstly reported in Ref. 16. The present sample contains three sets of 4 quantum wells, one at the center of the cavity and one in each Bragg mirror. The epitaxial layers (both cavity and mirrors) present a thickness gradient along the wafer so that the relative energy between the cavity mode and the exciton can be changed by moving the laser spot on the sample. We define the detuning as $\delta = E_C(k=0) - E_x(k=0)$, $E_C(k=0)$ and $E_x(k=0)$ being respectively the energy of the cavity mode and of the heavy hole (HH) exciton at in-plane wavevector $k=0$. Photoluminescence (PL) experiments are performed using a cw Ti:Sapphire laser focused onto a 50 μm diameter spot on the sample with a 30 mm focal lens and a 50° angle of incidence. The emission is collected through a 50 mm focal lens, angularly selected with a 0.5° angular resolution, spectrally dispersed with a double monochromator and detected with a Si avalanche photodiode. For all measurements, the cavity sample is

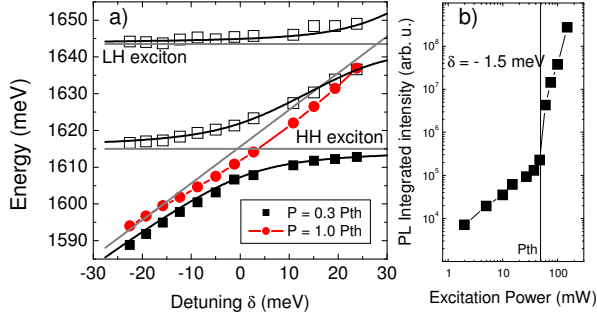


FIG. 1: (Color online) (a) squares : lower polariton branch energy measured by photoluminescence around normal incidence as a function of the cavity-exciton detuning δ , open squares: middle and upper polariton energy measured by photoluminescence excitation spectroscopy of the lower polariton as a function of δ , thick black line: fit of the three polariton branches, thin grey lines : deduced energy of the HH and LH exciton as well as of the uncoupled cavity mode; red circles : energy of the emission at threshold as a function of δ ; (b) Integrated intensity measured around normal incidence as a function of the non-resonant excitation power for $\delta = -1.5$ meV

held at 4K in a cold finger cryostat.

Fig.1(a) presents the energy of the lower polariton branch measured as a function of the detuning δ in the low density regime. The same graph also presents the energy of the middle and upper polaritons measured by photoluminescence excitation spectroscopy of the lower polariton branch. Two anticrossings are clearly observed between the cavity mode and the HH and light-hole (LH) exciton. The energy of the three polariton branches is well reproduced using a Rabi splitting of $\Omega_{HH} = 15$ meV and $\Omega_{LH} = 12.5$ meV with the HH and LH exciton respectively. Notice that the energy of the uncoupled cavity mode is precisely determined in the anti-crossing region by monitoring the spectral shift of the first reflectivity minimum on the high energy side of the mirror stop-band.

Photoluminescence measurements as a function of the excitation power were performed for various detunings under non-resonant excitation. For each detuning the laser energy is tuned to the first reflectivity minimum above the bragg-mirror stop-band, typically 120 meV above the polariton emission energy. A typical curve showing the power dependence of the integrated intensity measured at $k=0$ is shown in Fig. 1(b) for $\delta = -1.5$ meV. Above a threshold power P_{Th} , a very strong non-linear increase of the emission intensity is observed. To check whether the system stays in the strong coupling regime near threshold, we have performed angle resolved photoluminescence for different excitation powers. Fig. 2(a) presents photoluminescence spectra measured at low excitation power for several detection angles. The emission energy as a function of the deduced in-plane wavevector⁴ is summarized in Fig. 2(c). At low excitation power ($0.3 P_{th}$), the typical lower polariton dispersion

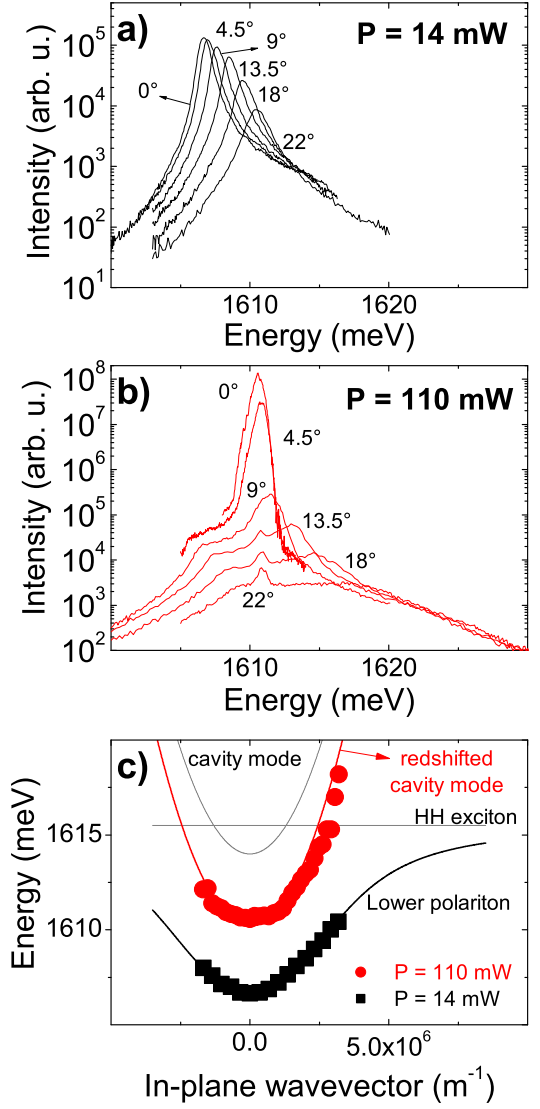


FIG. 2: (Color online) Photoluminescence spectra measured for different detection angles for an excitation power (a) $P = 14$ mW, (b) $P = 110$ mW; (c) Emission energy as a function of the in-plane wavevector (deduced from the detection angle) (squares) for $P = 14$ mW (circles), and $P = 110$ mW. The black line corresponds to the calculated lower polariton branch, the thin grey lines to the deduced uncoupled cavity mode and HH exciton, and the thick red line to the cavity mode redshifted by 3 meV. For the three figures, $\delta = -1.5$ meV

is observed. Fig. 2(b) shows PL spectra measured at the same point of the sample but for a higher excitation power ($2.4 P_{Th}$). At zero degrees, the emission spectrum presents a single intense line centered around 1611 meV. This line at 1611 meV is still visible at larger angle but with a reduced intensity: this is due to Rayleigh scattering within the sample. The emission spectra at finite detection angles present an additional emission line at higher energy than 1611 meV, continuously blueshifting with the angle of detection. The measured dispersion of

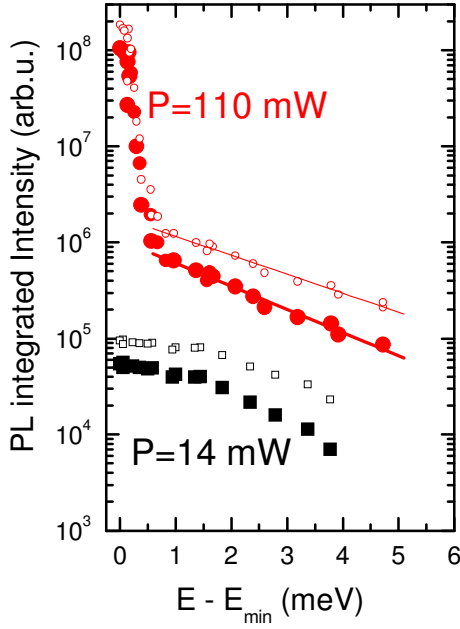


FIG. 3: (Color online) Full symbols: Photoluminescence integrated intensity as a function of the emission energy for (black squares) $P = 14\text{mW}$, (red circles) $P = 110\text{mW}$; Open symbols: Photoluminescence integrated intensity divided by the square of the polariton photonic Hopfield coefficient as a function of the emission energy for (open black squares) $P = 14\text{mW}$, (open red circles) $P = 110\text{mW}$; For each excitation power, E_{\min} corresponds to the emission energy measured at $k = 0$. $\delta = -1.5\text{meV}$

this line is summarized in Fig. 2(c). It is much steeper than the lower polariton branch at low excitation density. It can be well fitted using the cavity mode dispersion but rigidly red-shifted by roughly 3 meV. These angle resolved photoluminescence measurements show that the emission presents the energy dispersion of a pure photonic mode thus indicating that the strong coupling regime is lost for this range of excitation power.

Notice that the spectra of Fig. 2(b) also present a shoulder at energies smaller than 1611 meV, exhibiting a small blueshift with the angle of detection. This shoulder is due to polariton emission coming from the edge of the detection spot, where the carrier density is smaller and the strong coupling regime still exists. Indeed, given our angular resolution $\Delta\vartheta = 0.5^\circ$, the diameter d of the detection spot on the sample surface has a lower limit d_{\min} given by diffraction:

$$d_{\min} = 0.61 \frac{\lambda}{\tan(0.5 \cdot \Delta\vartheta)}$$

where λ is the emission wavelength in vacuum. This gives $d_{\min} = 110\text{ }\mu\text{m}$, which is two to three times the dimension of the excitation spot. This explains why polaritons in strong coupling regime coming from the edge of the excitation spot are also visible in the spectra of Fig. 2(b).

The energy of the emission line at threshold is plotted

as a function of detuning on Fig. 1(a). For strong positive or negative detunings, the emission energy at threshold matches that of the bare cavity mode deduced from fitting the polariton anticrossings. Nevertheless close to zero detuning or for positive detunings up to $+10\text{meV}$, the emission line at threshold lies at significantly lower energy than the calculated uncoupled cavity mode deduced from low density measurements. This reduced blueshift of the emission line when increasing excitation power could be mistaken for a sign that the system remains in the strong coupling regime, with a reduction of the Rabi splitting.

This incomplete blueshift, when lasing emission occurs, is clearly a feature occurring close to the excitonic resonance. This can be understood by considering the change of index of refraction of the GaAs when evolving from the excitonic regime toward the electron-hole plasma regime. A calculation of the low temperature GaAs index of refraction has been reported in ref. 17 for several carrier densities. At low carrier density, the real part of the index of refraction presents a derivative-shaped excitonic feature centered at the exciton resonance energy E_x , superimposed to a slowly varying function $\tilde{n}(E)$ slightly increasing with the energy. $\tilde{n}(E)$ accounts for the index of refraction due to all other resonances in the quantum well, and is the index of refraction value to be taken into account to calculate the energy of the uncoupled cavity mode. For high carrier densities, the excitonic feature progressively vanishes and the index of refraction $n_{HD}(E)$ presents a peak at an energy E_{HD} slightly higher than the exciton energy, superimposed to a slowly varying function close to $\tilde{n}(E)$. Thus for energies close to E_{HD} , $n_{HD}(E)$ is larger than $\tilde{n}(E)$. As a result, the cavity mode wavelength is at lower energy than what is deduced from low density measurements. Thus the overall emission blueshift with increasing excitation power is reduced. This incomplete blueshift occurs only near E_{HD} , i.e. for zero or positive detunings (see fig. 1a). It is induced by the refractive index of the QW layer only and is not influenced by the rest of the cavity layers. It is thus proportional to the overlap \mathcal{A} between the electromagnetic field of the cavity mode and the QW layer. Since the Rabi splitting is proportional to $\sqrt{\mathcal{A}}$, the spectral distance between the lasing emission and the calculated cavity mode varies as Ω^2 . It is thus particularly pronounced in our sample because it exhibits a very large Rabi splitting, as compared to samples of previous reports^{8,11} where the transition toward the weak coupling regime was studied.

Finally let us describe the emission pattern of the present microcavity. Fig. 3 summarizes the integrated intensity of the angle resolved measurements presented in Fig. 2. The integrated intensity is plotted as a function of the emission peak energy. In Fig. 3 we also plot with open symbols the integrated intensity divided by the square of the polariton Hopfield coefficient corresponding to the polariton photon content. In the strong coupling regime, this quantity is directly proportional to the po-

lariton population. At low excitation power, we find as in many previous works^{6,11} that the polariton population is not thermalized. This is because the polariton-polariton interaction time is much longer than the polariton lifetime. Above the lasing threshold in the weak coupling regime, the intensity distribution presents a pronounced peak close to $k=0$ (corresponding to $E = E_{min}$) and an exponential decay at higher energy. This shape remains very similar when dividing the intensity by the Hopfield coefficient (red open symbols) even if this operation is meaningless in the weak coupling regime. This emission pattern presents striking similarities with that reported for the polariton condensate^{6,15}. Such emission pattern has been highlighted as a proof that a thermalized condensate is achieved. This is an analogy to the case of atomic physics where a large fraction of the atoms is in the condensate, and coexists with a thermalized cloud of uncondensed atoms¹⁸. In our case, the system is in the photon lasing regime and the emission pattern probably simply reflects the energy distribution of the electron-hole pairs, the emission of which is filtered by the cavity mode.

Our aim in this paper is to underline that this emission pattern, a sharp peak near the energy minimum followed by a Boltzmann distribution at higher energies, can be similarly observed in a conventional laser in weak coupling regime. Although massive occupation of the lower energy states is an important characteristic of condensates, it cannot be used by itself to distinguish polariton condensation from photon lasing.

To conclude we have studied a III-V GaAs based semiconductor microcavity presenting a very small cavity volume and a large number of quantum wells, thus being an ideal candidate for Bose condensation. We confirm

as in previous reports that under high energy non resonant excitation, the polariton laser is not obtained and that only conventional photon lasing occurs. Nevertheless we show that because of refraction index changes when increasing the carrier density, this lasing regime can occur at much lower energy than the uncoupled cavity mode deduced from low density measurements. Thus in large Rabi splitting samples, a small emission blueshift or even the absence of any blueshift does not prove the persistence of the strong coupling. To demonstrate it one needs to monitor the polariton dispersion while being aware that a high angular resolution means that the emission from a large spatial area is collected. Great care should be taken in analyzing angular resolved PL in particular in spatially inhomogeneous geometries. Finally we want to underline that the emission pattern of the photon laser is the very same than that of a polariton condensate co-existing with a thermalized population of uncondensed polaritons.

In our opinion, an unambiguous proof for polariton condensation or polariton lasing is the observation of a second threshold at higher excitation density^{19,20,21} corresponding to photon lasing. This way it could be unambiguously shown that polariton condensation occurs at low excitation while at higher carrier density, the strong coupling regime vanishes and the onset of photon lasing is observed.

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